Scattering rates and lifetime of exact and boson excitons

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Abstract

Although excitons are not exact bosons, they are commonly treated as such provided that their composite nature is included in effective scatterings dressed by exchange. We here prove that, whatever these scatterings are, they cannot give both the exciton scattering rates T_{ij}^{-1} and lifetime τ_0 , correctly: A striking factor 1/2 exists between τ_0^{-1} and the sum of T_{ij}^{-1} 's, which originates from the composite nature of excitons, irretrievably lost when they are bosonized. This result, which appears as very disturbing at first, casts major doubts on the overall validity of bosonization for problems dealing with interacting excitons.

PACS.: 71.35.-y Excitons and related phenomena

Composite particles made of two fermions, like the excitons, are known to differ from exact bosons. It is however accepted that they can be treated as such, provided that their composite nature is included through effective scatterings dressed by exchange [1].

We here show that, in spite of the wide literature on bosonization [2] which claims to validate such a replacement, excitons can definitely differ from bosons, in physical effects linked to their interactions: Indeed, the lifetime τ_0 (due to exciton-exciton interaction) of the N-exciton state made with all excitons in their ground state 0, and the scattering rates $1/T_{ij}$ from this state towards states in which two excitons out of N are scattered to $(i, j) \neq (0, 0)$, are linked by

$$\tau_0^{-1} = \alpha \sum_{(i,j) \, couples} T_{ij}^{-1} ,$$
(1)

with $\alpha = 1$ if the excitons are bosonized, and $\alpha = 1/2$ if their composite nature is kept. This result proves that it is impossible to find effective scatterings for boson-excitons, giving *both* the lifetime and the scattering rates correctly.

The factor $\alpha = 1/2$ can be hard to accept at first because we could naïvely think that the probability to stay in the initial state plus the sum of probabilities to go to all other possible states must be equal to 1. As explained at the end of this letter, this naïve thinking in fact fails for excitons, due to a quite fundamental reason linked to their composite nature, irretrievably lost when they are bosonized.

Many works have been devoted to exciton-exciton (X-X) interactions [3-14]. They however [15] accept or end with an effective bosonic Hamiltonian, so that they do not tackle the consequences of transforming exact excitons into bosons. To study this problem, we have developed a new many-body theory [16-18], which allows to treat interactions between composite bosons, all previous many-body theories dealing with true fermions or true bosons only. The case of composite excitons is far more complex. (i) The carriers being indistinguishable, there is no way to know if a given exciton is made of (e, h) or (e', h). (ii) Due to this uncertainty, there is no way to extract from the Coulomb terms of the semiconductor Hamiltonian H, the part corresponding to a potential V_{XX} between exciton operators. With H not written as $H_X + V_{XX}$, perturbation theory, which is the standard way to solve problems dealing with interactions, cannot be used anymore. (iii) Far worse, excitons feel each other not only through Coulomb interaction, but also through Pauli exclusion. This "Pauli interaction", which originates from statistical departure, can

a priori exist in the absence of any Coulomb process. It is the conceptually new part of our many-body theory. It requires a very peculiar procedure to have it appearing.

As most readers are not yet familiar with this new theory [16-18], let us recall its main points. Two kinds of "scatterings" between excitons, instead of one, appear in this theory: (i) ξ_{mnij}^{dir} , which has the dimension of an energy, corresponds to direct Coulomb processes between both the "in" excitons (i, j) and the "out" excitons (m, n). (ii) λ_{mnij} , which is dimensionless, comes from "Pauli interaction", without any Coulomb contribution. In ξ_{mnij}^{dir} , the "in" and "out" excitons are made with the same pairs, while in λ_{mnij} they just exchange their electrons (eqs. (26,28) of ref. [16]),

$$\xi_{mnij}^{\text{dir}} = \frac{1}{2} \int d\mathbf{r}_e \, d\mathbf{r}_{e'} \, d\mathbf{r}_h \, d\mathbf{r}_{h'} \, \phi_m^*(\mathbf{r}_e, \mathbf{r}_h) \, \phi_n^*(\mathbf{r}_{e'}, \mathbf{r}_{h'}) \, \left[V_{ee'} + V_{hh'} - V_{eh'} - V_{e'h} \right]$$

$$\times \phi_i(\mathbf{r}_e, \mathbf{r}_h) \, \phi_j(\mathbf{r}_{e'}, \mathbf{r}_{h'}) + (m \leftrightarrow n) \,, \tag{2}$$

$$\lambda_{mnij} = \frac{1}{2} \int d\mathbf{r}_e d\mathbf{r}_{e'} d\mathbf{r}_h d\mathbf{r}_{h'} \phi_m^*(\mathbf{r}_{e'}, \mathbf{r}_h) \phi_n^*(\mathbf{r}_e, \mathbf{r}_{h'}) \phi_i(\mathbf{r}_e, \mathbf{r}_h) \phi_j(\mathbf{r}_{e'}, \mathbf{r}_{h'}) + (m \leftrightarrow n) . \quad (3)$$

 $\phi_i(\mathbf{r}_e, \mathbf{r}_h) = \langle \mathbf{r}_e, \mathbf{r}_h | B_i^{\dagger} | v \rangle$ is the wave function of the i exciton at hand (Wannier, Frenkel, any type). B_i^{\dagger} , which creates this i exciton, is such that $(H - E_i)B_i^{\dagger} | v \rangle = 0$, where H is the semiconductor Hamiltonian, made of the electron and hole kinetic energies plus the e-e, h-h, e-h Coulomb interactions. For bound states, we can show that $\lambda_{mnij} \simeq \mathcal{V}_X/\mathcal{V}$ and $\xi_{mnij}^{\text{dir}} \simeq R_X \mathcal{V}_X/\mathcal{V}$, with \mathcal{V} , \mathcal{V}_X and R_X being the sample volume, the exciton volume and the exciton Rydberg.

Due to the composite nature of the excitons, we have (eq. (7) of ref. (16)),

$$G_{ijmn} = \langle v | B_i B_j B_m^{\dagger} B_n^{\dagger} | v \rangle = \delta_{im} \delta_{jn} + \delta_{in} \delta_{jm} - 2\lambda_{ijmn} , \qquad (4)$$

which shows that the exciton states are non-orthogonal, while (eq. (5) of ref. (16)),

$$B_i^{\dagger} B_j^{\dagger} = -\sum_{mn} \lambda_{mnij} B_m^{\dagger} B_n^{\dagger} , \qquad (5)$$

which shows that the exciton states form an overcomplete set. (Let us recall that for boson-excitons, the Pauli scatterings λ_{mnij} reduce to zero).

Any matrix element between N-exciton states can be calculated in terms of ξ_{mnij}^{dir} and λ_{mnij} . A major difficulty however remains if we want to work with excitons: While for boson-excitons, the exact Hamiltonian H is replaced by $H_X + V_{XX}$, so that we can use the Fermi golden rule to get the lifetime and scattering rates, there is no such a V_{XX}

for exact excitons, so that we first have to construct unconventional expressions of these quantities in which V does not appear.

Lifetime and scattering rates in terms of H

The time evolution of an initial state $|\psi_0\rangle$ can be written as

$$|\psi_t\rangle = e^{-i\hat{H}t}|\psi_0\rangle = |\psi_0\rangle + |\tilde{\psi}_t\rangle ,$$
 (6)

where $\hat{H} = H - \langle H \rangle$, with $\langle H \rangle = \langle \psi_0 | H | \psi_0 \rangle$. This gives

$$|\tilde{\psi}_t\rangle = F_t(\hat{H})\,\hat{H}|\psi_0\rangle ,$$
 (7)

 $F_t(E) = (e^{-iEt}-1)/E$ being such that $|F_t(E)|^2 = 2\pi t \, \delta_t(E)$, where $\delta_t(E) = (\pi E)^{-1} \sin(Et/2)$ is the usual delta function of width (2/t).

The $|\psi_0\rangle$ lifetime, defined as $|\langle \psi_0 | \psi_t \rangle|^2 \simeq 1 - t/\tau_0$, then reads $t/\tau_0 \simeq -(\langle \psi_0 | \tilde{\psi}_t \rangle + c.c)$, which is nothing but $\langle \tilde{\psi}_t | \tilde{\psi}_t \rangle$, due to $\langle \psi_t | \psi_t \rangle = 1$. This leads to

$$t/\tau_0 \simeq \langle \psi_0 | \hat{H} | F_t(\hat{H}) |^2 | \hat{H} | \psi_0 \rangle . \tag{8}$$

As for the scattering rate from $|\psi_0\rangle$ to an arbitrary state $|\phi_n\rangle$, defined as $t/T_n \simeq |\langle\phi_n|\tilde{\psi}_t\rangle|^2$ [19], it reads, due to eq. (7)

$$t/T_n \simeq \left| \langle \phi_n | F_t(\hat{H}) \, \hat{H} | \psi_0 \rangle \right|^2 .$$
 (9)

These unconventional expressions of τ_0 and T_n do not contain any V as required. From them, it is however easy to recover the conventional ones, since, for $H = H_0 + V$ and $|\psi_0\rangle = |0\rangle$ with $(H_0 - \mathcal{E}_n)|n\rangle = 0$, we have $\hat{H} |\psi_0\rangle = \sum_{n\neq 0} |n\rangle \langle n|V|0\rangle$.

Exact excitons

We first consider the lifetime and scattering rates of exact excitons. Let us take as initial state, $|\psi_0\rangle = B_0^{\dagger N} |v\rangle / \sqrt{N! F_N}$, where B_0^{\dagger} is the creation operator for one exciton in its ground state, while F_N is such that $N! F_N = \langle v | B_0^N B_0^{\dagger N} | v \rangle$ for $|\psi_0\rangle$ to be normalized [20]. This state can be seen as the one coupled to N identical photons [21] tuned on the ground state exciton, before its relaxation to the N-pair ground state — otherwise this initial state would not change with time.

The reader having difficulties with the various factors N appearing in this work, can just take N=2. The N-exciton problem then reduces to a far simpler 2-exciton problem, which still has the striking factor $\alpha=1/2$ appearing. The various N's of the final results

are actually the same for exact and boson excitons, as physically expected: A difference in these N dependences induced by the composite nature of the exciton, would be even harder to accept than the factor $\alpha = 1/2$!

To get $\hat{H} | \psi_0 \rangle$ appearing in eqs. (8,9), we use our new theory. We first calculate $\left[H, B_0^{\dagger N} \right]$ by induction from eqs. (14,20) of ref. [18], namely $\left[H, B_i^{\dagger} \right] = E_i B_i^{\dagger} + V_i^{\dagger}$ and $\left[V_i^{\dagger}, B_j^{\dagger} \right] = \sum_{mn} \xi_{mnij}^{\text{dir}} B_n^{\dagger} B_n^{\dagger}$, where, due to eq. (5), ξ_{mnij}^{dir} can be replaced by $(-\xi_{mnij}^{\text{in}})$, or better by

$$\hat{\xi}_{mnij} = (\xi_{mnij}^{\text{dir}} - \xi_{mnij}^{\text{in}})/2 = -\sum_{rs} \lambda_{mnrs} \hat{\xi}_{rsij} . \tag{10}$$

The "in" exchange Coulomb scattering, defined as $\xi_{mnij}^{\text{in}} = \sum_{rs} \lambda_{mnrs} \xi_{rsij}^{\text{dir}}$, reads as ξ_{mnij}^{dir} given in eq. (2), with $\phi_m^*(\mathbf{r}_e, \mathbf{r}_h) \phi_n^*(\mathbf{r}_{e'}, \mathbf{r}_{h'})$ replaced by $\phi_m^*(\mathbf{r}_{e'}, \mathbf{r}_h) \phi_n^*(\mathbf{r}_e, \mathbf{r}_{h'})$, so that, using eq. (3), we have $\sum_{rs} \lambda_{mnrs} \xi_{rsij}^{\text{in}} = \xi_{mnij}^{\text{dir}}$. From the obtained $[H, B_0^{\dagger N}]$, we find

$$\hat{H}B_0^{\dagger N}|v\rangle = (1/2)N(N-1)B_0^{\dagger N-2} \left[\Delta_0 B_0^{\dagger 2} + \sum_{mn \neq 00} \hat{\xi}_{mn00} B_m^{\dagger} B_n^{\dagger} \right] |v\rangle , \qquad (11)$$

with $\Delta_0 = \hat{\xi}_{0000} + [NE_0 - \langle H \rangle]/[N(N-1)/2]$. Using $\langle H \rangle$ calculated in ref. [22], this Δ_0 actually reads $\hat{\xi}_{0000}(-1 + O(\eta))$, so that $\hat{H}B_0^{\dagger N}|v\rangle$ is first order in Coulomb scattering, as physically expected.

We can then use eq. (5) to rewrite $\Delta_0 B_0^{\dagger 2}$ as a sum of $\Delta_0 \lambda_{mn00} B_m^{\dagger} B_n^{\dagger}$. Since these terms are one Pauli scattering smaller than the $\hat{\xi}_{mn00} B_m^{\dagger} B_n^{\dagger}$ terms of the sum appearing in eq. (11), we can, at lowest order in the exciton interactions, drop $\Delta_0 B_0^{\dagger 2}$ in front of the sum of eq. (11), so that at this order, $\hat{H} B_0^{\dagger N} |v\rangle$ is made of states with two excitons (m, n) outside 0.

To get the transition rate T_{ij}^{-1} from $|\psi_0\rangle$ to one of these states $|\phi_{ij\neq 00}\rangle = a_{ij}B_0^{\dagger N-2}B_i^{\dagger}B_j^{\dagger}|v\rangle$, with a_{ij} being a normalization factor, we make $F_t(\hat{H})$ acting on the left in eq. (9). Since $\hat{H}|\psi_0\rangle$ is first order in the interactions already, we can replace $F_t(\hat{H})$ by its free-exciton contribution, $F_t(E_i + E_j - 2E_0)$. We are left with the scalar product of N-exciton states. For N=2, it is just G_{ijmn} given in eq. (4), while in the large N limit, and for $(i,j,m,n)\neq 0$, we find [23]

$$\langle v|B_iB_iB_0^{N-2}B_0^{\dagger N-2}B_m^{\dagger}B_n^{\dagger}|v\rangle \simeq G_{iimn}(N-2)!F_{N-2}$$
 (12)

Since, due to eqs. (4,10), $\sum_{mn} G_{ijmn} \hat{\xi}_{mn00} = 4\hat{\xi}_{ij00}$, eqs. (9-12) lead to

$$T_{i\neq j}^{-1} \simeq t^{-1}(1/4)N(N-1) \left| F_t(E_i + E_j - 2E_0) \sum_{mn\neq 00} G_{ijmn} \hat{\xi}_{mn00} \right|^2$$

$$\simeq 2\pi N(N-1) |\xi_{ij00}^{\text{dir}} - \xi_{ij00}^{\text{in}}|^2 \delta_t(E_i + E_j - 2E_0) , \qquad (13)$$

the transition rate to $|\phi_{ii}\rangle$ being zero, due to energy and momentum conservation.

If we now turn to the $|\psi_0\rangle$ lifetime (eq. (8)), we find, in the same way, that it reads [24]

$$\tau_0^{-1} \simeq t^{-1} (1/4) N(N-1) \sum_{0 \neq i \neq j \neq 0} |F_t(E_i + E_j - 2E_0)|^2 \hat{\xi}_{00ij} \sum_{mn \neq 00} G_{ijmn} \hat{\xi}_{mn00}$$

$$\simeq \frac{1}{4} \sum_{0 \neq i \neq j \neq 0} T_{ij}^{-1} = \frac{1}{2} \sum_{(i,j) couples} T_{ij}^{-1} , \qquad (14)$$

as claimed in eq. (1).

Boson excitons

Bosonizing the excitons corresponds to replace the exact exciton operators B_i by \bar{B}_i 's, with $[\bar{B}_i, \bar{B}_j^{\dagger}] = \delta_{ij}$, and the exact Hamiltonian H by $H_X + V_{XX}$, with $H_X = \sum_i E_i \bar{B}_i^{\dagger} \bar{B}_i$ and $V_{XX} = (1/2) \sum_{mnij} \xi_{mnij}^{\text{eff}} \bar{B}_m^{\dagger} \bar{B}_i \bar{B}_j$. This leads to replace the G_{ijmn} of eq. (4) by $\bar{G}_{ijmn} = \delta_{im}\delta_{jn} + \delta_{in}\delta_{jm}$ and the $\hat{\xi}_{mn00}$ of eq. (11) by ξ_{mn00}^{eff} . In the same way, we are led to bosonize the initial state by taking it as $|\bar{\psi}_0\rangle = \bar{B}_0^{\dagger N}|v\rangle/\sqrt{N!}$. We then find that the first lines of eqs. (13-14) are still valid with $\sum_{mn} \bar{G}_{ijmn} \xi_{mn00}^{\text{eff}}$ now equal to $2\xi_{ij00}^{\text{eff}}$. (Note that the similar sum for exact excitons gives $4\hat{\xi}_{ij00}$: This is actually the mathematical reason for the factor 2 change between the α 's of eq. (1)). This leads us to find

$$\bar{T}_{i\neq j}^{-1} \simeq 2\pi N(N-1) |\xi_{ij00}^{\text{eff}}|^2 \delta_t(E_i + E_j - 2E_0) ,$$
 (15)

$$\bar{\tau}_0^{-1} \simeq \frac{1}{2} \sum_{0 \neq i \neq j \neq 0} \bar{T}_{ij}^{-1} = \sum_{(i,j) couples} \bar{T}_{ij}^{-1} ,$$
 (16)

which is just what the usual Fermi golden rule readily gives.

In order to grasp why exact and boson excitons can have a different α , let us note that the link between $\bar{\tau}_0^{-1}$ and the \bar{T}_{ij}^{-1} 's can be recovered, without calculations, just from the closure relation between bosons. This relation reads

$$1 = (1/N!) \sum_{i_1, \dots, i_N} \bar{B}_{i_1}^{\dagger} \cdots \bar{B}_{i_N}^{\dagger} |v\rangle \langle v|\bar{B}_{i_N} \cdots \bar{B}_{i_1}$$

$$= |\bar{\psi}_0\rangle \langle \bar{\psi}_0| + \sum_{i \neq 0} |\bar{\phi}_i\rangle \langle \bar{\phi}_i| + \frac{1}{2} \sum_{(i,j) \neq 0} (1 + \delta_{ij}) |\bar{\phi}_{ij}\rangle \langle \bar{\phi}_{ij}| + \cdots .$$

$$(17)$$

where $|\bar{\phi}_i\rangle$, $|\bar{\phi}_{ij}\rangle$, ... are the *normalized* boson-exciton states with one, two, ... boson-excitons outside 0. By inserting eq. (17) into $1 = \langle \bar{\psi}_t | \bar{\psi}_t \rangle$, we get, to lowest order in the

interactions,

$$1 \simeq |\langle \bar{\psi}_0 | \bar{\psi}_t \rangle|^2 + (1/2) \sum_{0 \neq i \neq j \neq 0} |\langle \bar{\phi}_{ij} | \bar{\psi}_t \rangle|^2, \tag{18}$$

as energy and momentum conservations impose $\langle \bar{\phi}_i | \bar{\psi}_t \rangle = 0 = \langle \bar{\phi}_{ii} | \bar{\psi}_t \rangle$ for $i \neq 0$. The first term of eq. (18) is just $1 - t/\bar{\tau}_0$, while $\left| \langle \bar{\phi}_{ij} | \bar{\psi}_t \rangle \right|^2 = t/\bar{T}_{ij}$, so that eq. (16) readily follows from eq. (18).

Discussion

By comparing eqs. (13) to eq. (15), we see that the transition rates of exact and boson excitons are equal if we enforce $\xi_{mnij}^{\text{eff}} = \xi_{mnij}^{\text{dir}} - \xi_{mnij}^{\text{in}}$. This scattering differs from the one used up to now [1], namely $\tilde{\xi}_{mnij}^{\text{eff}} = \xi_{mnij}^{\text{dir}} - \xi_{mnij}^{\text{out}}$, with $\xi_{mnij}^{\text{out}} = (\xi_{ijmn}^{\text{in}})^*$. We have however shown [16] that, as $\xi_{mnij}^{\text{out}} \neq (\xi_{ijmn}^{\text{out}})^*$, this $\tilde{\xi}_{mnij}^{\text{eff}}$ is physically unacceptable because the corresponding Hamiltonian is non hermitian. The new ξ_{mnij}^{eff} has also to be rejected for the same reason.

The most striking result of this letter is however not so much the difficulty in finding physically reasonable effective scatterings giving the exact scattering rates, but the link between the inverse lifetime and the sum of scattering rates: The additional factor 1/2 found for exact excitons actually invalidates the overall validity of bosonization, because it is not possible to find a set of ξ_{mnij}^{eff} 's giving both, the lifetime and the scattering rates, correctly.

As the exact-exciton calculations rely on a theory which is not yet commonly used, the reader can reasonably question the correctness of this striking result which appears as very disturbing at first. It is however easy to be, at least, convinced that different links between τ_0^{-1} and the sum of T_{ij}^{-1} 's for exact and boson excitons are quite reasonable: As shown above, the link for boson-excitons comes from the closure relation which exists between boson-exciton states, these states forming an orthogonal basis. On the opposite, as clear for N=2 already (see eqs. (4,5)), the basis made of exact-exciton states is not only non-orthogonal but, far worse, overcomplete. Consequently, the closure relation for boson excitons cannot be transposed to exact excitons, just as the link between τ_0^{-1} and the T_{ij}^{-1} 's. Since this overcompleteness of the exciton state set physically comes from the exciton composite nature, the additional factor 1/2 found between τ_0^{-1} and the sum of T_{ij}^{-1} 's has to be seen as a non trivial signature of the fact that excitons are deeply made of two fermions. Our letter shows in a striking way that this composite nature can be

crucial for problems dealing with interacting excitons, in spite of a widely spread belief.

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